



MECHANISTIC STUDY OF CO₂ ADSORPTION ON SOLID AMINE SORBENT

Background

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The capture of CO₂ from gas mixture can be achieved by using solvents, cryogenic techniques, membranes, and solid sorbents. Amine-based, wet scrubbing systems have been used as capture techniques for CO₂ removal from flue gas streams. The possible reaction sequences in an aqueous system using primary and secondary alkanolamines reacting with dissolved CO₂ are shown in Figure 1.

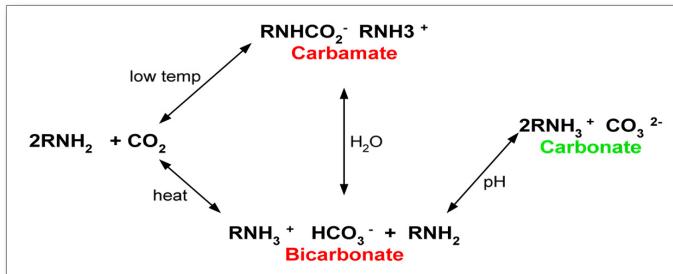


Figure 1. Possible reaction sequence for the capture of carbon dioxide by liquid amine-based systems.

According to Figure 1, the majority of the CO₂ captured will result in the formation of bicarbonate in these liquid amine capture systems. In aqueous media, there is a requirement of 2 moles of amine/ mole of CO₂ for the formation of stable bicarbonate compounds resulting in the capture of CO₂. For the case of solid amine sorbents, the detail reaction sequence is unclear thus far.

One strategy to develop a high CO₂ adsorption capacity solid sorbent is to graft amine functional group(s) containing species on the high surface area supports. A high surface area support will allow us to graft a higher number of active amine sites per gram of support. The nature of the grafted amine functional groups (primary or secondary amine) will determine the amount of CO₂ adsorbed and the energy required for regenerating the sorbent. The interaction between the CO₂ and NH/NH₂ groups is expected to be stronger than van der Waals forces but weaker than a covalent/ionic bond such that the CO₂ bound to the amino groups can be released with temperature swing. In order to fabricate a high CO₂ capture capacity solids amine sorbent, we must understand the fundamental interactions between CO₂ and amine on solid surface.



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Primary Project Goal

The goal of the research is to determine what factors affect the efficiency of the solid CO₂ sorbents.

Objectives

- Use infrared (IR) spectroscopy to investigate the mechanism of CO₂ adsorption/desorption on amine sorbents;
- Estimate the effect of moisture content on the adsorption kinetics and sorption capacities of amine sorbents.

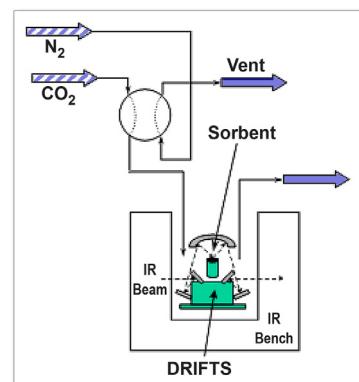


Figure 2. An assembled apparatus included a diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) cell for sorption/desorption measurements.

Accomplishments

IR study of the interactions between CO₂ and amines:

The experimental work for carbon dioxide interaction with 3-propylamine functionalized gel was initiated by using infrared spectroscopy. Infrared spectra of an Aldrich 36,425-8 sample and 3-propylamine functionalized silica gel were grafted on mesoporous SBA-15 particles which were collected repeatedly using two complete regeneration cycles. The cycles were collected at different partial pressures of carbon dioxide in nitrogen and demonstrated a fairly reproducible correlation between the free CO₂ gas concentration and the intensity of the signal corresponding to various adsorbed species. The amount of the weakly bound bidentate bicarbonate was directly related to the partial pressure of CO₂. Some carbonate complexes were not easily removed, even at 90 °C. Carbamate formation becomes a dominant mechanism of adsorption on 3-propylamine at higher CO₂ pressures.

Procedure:

- Prepare Aldrich 36,425-8 sample, 20 mg for FTIR analysis
- Outgas in N₂ at 90 °C for 4 hrs.
- Ambient T, N₂
- Ambient T, Switch to CO₂
- Ambient T, Switch to N₂ to remove CO₂
- Repeat cycle with regenerated sample

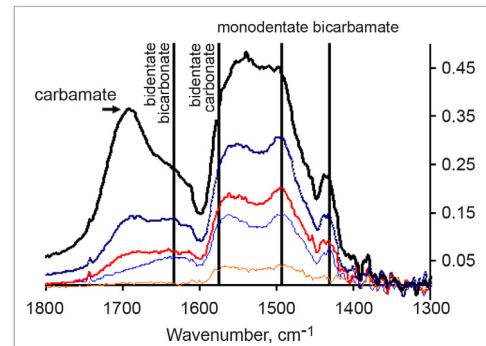


Figure 3. The amount of the weakly bound bicarbonates is directly related to the partial pressure of CO₂. Some carbonate complexes are not as easily removed, even at 90 °C. Carbamate formation becomes a dominant mechanism of adsorption on 3-propylamine at high CO₂ pressures.

Benefits

The separation process in aqueous amines is effective for carbon dioxide adsorption but it presents a corrosion and degradation problem involving extensive energy use for regeneration of the amine solution. The solid sorbent, in addition to elimination of these problems, may offer an advantage of high surface area density available for adsorption. This may also enhance the fundamental knowledge of CO₂-amine interactions on solid sorbents.